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Leaching of radionuclides from activated soil into groundwater

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ABSTRACT

Soil samples collected from the CERN site were irradiated by secondary radiation from the 400 GeV/c SPS proton beam at the H4IRRAD test area. Water samples were also irradiated at the same time. Detailed gamma spectrometry measurements and water scintillation analysis were performed to measure the radioactivity induced in the samples. FLUKA Monte Carlo simulations were performed to benchmark the induced radioactivity in the samples and to estimate the amount of tritium produced in the soil. Two leaching procedures were used and compared to quantify the radioactivity leached by water from the activated soil. The amount of tritium coming from both the soil moisture and the soil bulk was estimated. The present results are compared with literature data for the leaching of ^{3}H and ^{22}Na.

1. Introduction

Accelerators and experimental facilities are typically sited either underground or at grade with thick concrete walls and substantial earth berms to provide cost-effective shielding. Radiation interacts with the shielding materials generating induced radioactivity in the concrete or earth. In general, the induced radioactivity remains confined in the shield material. However, some activation may occur outside the accelerator enclosure, primarily in adjacent groundwater and soil. If the production of radioactive nuclides in the accelerator structure and in the concrete walls of the accelerator room is a concern for the personnel, the radionuclides created in the groundwater or in the earth represent a collective danger, because they may be transported into the environment. This was the case, for example, of the tritium leak accident discovered at Brookhaven National Laboratory (BNL) in 1997, when a considerable amount of tritium leached out of the concrete walls of a reactor's spent fuel pool. The laboratory analysis of water samples taken near the reactor revealed concentrations of tritium that greatly exceeded the EPA (US Environmental Protection Agency) drinking water standards. Shortly after the tritium levels were made public, a firestorm of public concern blew up and caused the contractor dismissal at the BNL (United States General Accounting Office, 1997).

Several authors investigated the radioisotope production in earth and water, either experimentally or via Monte Carlo simulations (Nelson, 1965; Hoyer, 1968; Warren et al., 1969; Ranft and Goebel, 1970; Thomas, 1970; Stapleton and Thomas, 1972; Rindi, 1972; Borak et al., 1972; Baker, 1975, 1985; Sullivan, 1987; Malensek et al., 1993; Baker et al., 1997; Tesch, 1997; Racky et al., 1998; Wehmann and Childress, 1999; Vincke and Stevenson, 2000; Rokni et al., 2000; Agosteo et al., 2005; Vogt et al., 2008). They describe and examine the most important isotopes of concern, but few studies have addressed the problem of the radionuclide migration from the activated soil to the groundwater. Remarkable exceptions are represented by the work of Borak et al. (1972) and Baker et al. (1997). They performed laboratory measurements of the production, leaching and movement of radionuclides produced in soil samples. Most of the later papers (Baker, 1975, 1985; Sullivan, 1987; Malensek et al., 1993; Baker et al., 1997; Tesch, 1997; Racky et al., 1998; Wehmann and Childress, 1999; Vincke and Stevenson, 2000; Rokni et al., 2000) directly or indirectly refer to the results of Borak and Baker for the part concerning the leaching of radioactivity.

This paper investigates the leaching of radioactivity in the type of soil ("molasse") of the CERN region, and is part of the preventive studies conducted by the Laboratory to be prepared for a potential mishap. The scope of this study is to provide experimental data in order that, should an incident occur, transfer of radioactivity to the soil can be ready assessed. This is achieved by assessing the radionuclide production from secondary radiation in earth and water and by quantifying the activity concentration of the

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radionuclides leaching into the groundwater. The study mainly focuses on ^{3}H and ^{22}Na, which are the longest-lived radionuclides and the most problematic from a radiation protection point of view. The chemical composition of the soil and the activation experiment are described. The results of the gamma spectrometry and liquid scintillation analyses are discussed. FLUKA Monte Carlo (Ferrari et al., 2005; Battistoni et al., 2007) simulations were performed to estimate the amount of tritium in the soil. In order to validate the FLUKA capability in predicting the production of radioactivity in this specific context, the simulation results were benchmarked against the measured radionuclides. Two alternative approaches were investigated: 1) the mixing system, in which water is mixed with the soil and 2) the flowing system, in which the water flows through the soil. The results of both leaching procedures are analysed and compared. The amount of tritium coming from the soil moisture and the soil bulk was also estimated, a novel result with respect to previous studies.

2. Soil and water samples

The soil samples used for the irradiation were collected on the Prévessin site of CERN, at a depth of 24 m. CERN is located in the Geneva basin, which is filled by sedimentary deposits called molasse. The chemical analysis of the molasse rock was carried out by the EMPA laboratory in Dübendorf (Switzerland) via X-ray fluorescence spectrometry (WD-XRF). Soil samples were dried before the XRF measurements. The XRF analysis is not reliable in quantifying the 1st and 2nd period elements (from H to Ne) in inorganic solid matrices (EMPA, 2011). For this reason, the oxygen and carbon contents were extrapolated using typical values of molasse soil (Vincke and Stevenson, 2000). The results of this chemical assay are shown in Table 1.

The knowledge of the natural water content of the soil (moisture) is essential to discriminate the fraction of radioactivity leaching into groundwater from the soil, from the fraction coming from the moisture. The moisture was measured in CERN Environmental laboratory by drying a known amount of earth. The soil moisture content may be expressed by weight as the ratio of the mass of water present to the dry weight of the soil sample. To determine this ratio, the samples and container were weighed in the laboratory both before and after drying, the difference being the mass of water originally in the sample. The water content measured in the soil specimen was 5% by weight.

Three cylindrical plastic containers 5 cm long and 4.5 cm in diameter were filled with 100 g of soil. A sample of drinking water was collected in a 50 ml plastic container 10 cm long and 2.5 cm in diameter (Fig. 1). The natural radioactivity in the samples was measured before irradiating them in the experiment. A sample of molasse was measured with a Ge detector for 5×10^{4} s (about 14 h). The main contribution of the natural radioactivity comes from ^{40}K (0.32 Bq g^{-1}) with small contributions from the thorium (0.018 Bq g^{-1}) and uranium chains (0.019 Bq g^{-1}). These results are in good agreement with the activity concentrations given in Vincke and Stevenson (2000). Additional information on the samples and on the chemical analysis can be found in La Torre et al. (2012a).

Table 1
Chemical composition of dried soil sample (density: ~1.4 g cm^{-3}).

Element	O	Si	Ca	Al	C	Fe	Mg	K	Na	Ti
(g/100 g)	38.8[a]	24	16	6.8	5[a]	4	2	1.9	0.7	0.42
	Mn	Ba	P	Sr	Zn	Cr	Zr	Eu	Ni	S
(g/100 g)	0.11	0.06	0.06	0.05	0.03	0.02	0.02	0.01	0.01	0.01

[a] Extrapolated value, not quantifiable by XRF analysis.

Fig. 1. Soil and water samples used in the activation experiment.

3. Irradiation set-up

The activation experiment was carried out at the H4IRRAD facility in the North Experimental Area of the CERN Super Proton Synchrotron (SPS). This facility hosts a copper target struck by the SPS primary proton beam with momentum of 400 GeV/c and average intensity of 3×10^{9} protons per pulse (over a supercycle of about 45 s and an extraction length of ~5 s). The soil and water containers were installed under the copper target. A schematic view of the experimental set up is shown in Fig. 2. The activation experiment was carried out at the H4IRRAD facility (Biskup et al., 2011) in CERN North Experimental Area. The soil and water containers were installed under the copper target struck by the SPS primary proton beam with momentum of 400 GeV/c and average intensity of 3×10^{9} protons per pulse (over a supercycle of about 45 s and an extraction length of ~5 s). An argon ionisation chamber (XION) placed in the H4 beam line just upstream of the copper target monitored the intensity of the primary beam. One XION-count corresponds to (6900 ± 690) particles impinging on the target (La Torre et al., 2012b).

3.1. Analysis of the irradiated samples

Immediately after irradiation with ~ 7.5×10^{13} accumulated protons, the dose rate of the samples was of the order of a few mSv/h. Most of this radioactivity was due to very short half-life radioisotopes. Since the radioisotopes of interest to this study have medium and long half-life, the samples were let decaying for 10 days before counting. The earth and water samples were measured with a high sensitivity, low-background, high-purity germanium (HPGe) detector by Canberra. The data acquisition and analysis was carried out using Canberra's Genie-2000 spectrometry software and the PROcount-2000 counting procedure software (Genie-2000 Software, 2000). This is a comprehensive software package for data acquisition, display and analysis, which includes a set of advanced spectrum analysis algorithms providing a complete analysis of gamma ray spectra. Three gamma spectrometry analyses were performed for each sample, at cooling times of one week, one month and two months.

The soil samples could not be directly counted for ^{3}H due to the low beta-particle endpoint energy (18 keV), which is absorbed in the sample. For this reason, the tritium activity in the soil was estimated via Monte Carlo calculations (Section 3.2). The tritium activity in the water was determined using a liquid scintillation counter (Packard TRI-CARB 3180TR/SL), measuring a mixture of 8 ml of activated water and 12 ml of so-called liquid scintillation cocktail (Packard Ultima Gold LLF). In case of high precision